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## (54) POLYLACTIC ACID LAMINATED BIAXIALY STRETCHED FILM

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To provide a polylactic acid laminated biaxially stretched film having heat salability and sufficient heat shrinkage resistance.

**SOLUTION:** This polylactic acid laminated biaxially stretched film comprises at least two layers based on a polylactic acid polymer. A relation between the D-lactic acid containing ratio Da (%) of the crystalline polylactic acid polymer constituting one layer of the laminated film and the D-lactic acid containing ratio Db (%) of the polylactic acid polymer constituting the other one layer of the laminated film is  $Da \leq 7$  and  $Db - Da \geq 3$ , and the other one layer is constituted of at least one outermost layer of the laminated film.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the polylactic acid system laminating biaxial oriented film of the good biodegradability of heat-sealing nature.

[0002]

[Description of the Prior Art] Conventionally, cellophane was widely used as a film for a package, and was used for the package of a fiber package, confectionery, etc., the envelope for drug, etc., and was used as a film which carried out the coat of the polyvinylidene chloride to the front face, and has dampproofing and heat-sealing nature. Since this cellophane is using as the main raw material quality of pulp which can be taken from wood, it is the description that there is resolvability, but since the manufacture approach called casting method produced is taken once carrying out the chemical treatment of the quality of pulp and dissolving, productivity is low and needs investment which prepares a facility from the point of waste water treatment. For this reason, most has replaced the polyethylene which consists of a petroleum origin raw material with a low manufacturing cost, and the film which consists of polyolefine or aromatic polyester by the end of today. The film which gave thermal resistance, shrinkage characteristics, heat-sealing nature, printing nature, dampproofing, fog resistance, etc. by the processing method with various polyethylene, polyolefines, and aromatic polyester of this petroleum origin is manufactured, and the application is also widely used also not only as a packing material but as industrial material.

[0003] However, the above-mentioned cellophane has many amount used, though there is a property of not sorbing the scent of a perfume, tea, coffee, etc., etc., it is improved as a packing material and it has the above-mentioned trouble. Moreover, the film which consists of a petroleum origin raw material has much calorific value at the time of combustion, and has a possibility of damaging a combustion furnace during combustion processing. furthermore -- although reclamation processing is carried out in many cases -- the -- for scientific and living thing-stability, it hardly decomposed, but it remained and problems, such as shortening the life of reclaimed ground, are caused. For this reason, like cellophane, among soil, what is decomposed underwater is desired and many researches are made.

[0004] Polylactic acid is raised as an example of the ingredient with which development is furthered today and which is underwater disassembled among soil, i.e., a biodegradability ingredient. The amount of heat of combustion is below one half of polyethylene, and hydrolysis advances automatically by the inside of soil, and underwater one, and, subsequently this polylactic acid serves as a harmless decomposition product by the microorganism. A moldings and the research which specifically obtains containers, such as a film, a sheet, and a bottle, etc. are made using current and polylactic acid.

[0005] By the way, elongation has only several % and the non-oriented film of polylactic acid is a weak ingredient. For this reason, a thin non-extended film is impractical as an object for a package. It is already well-known that the high film of the practicality which controlled heat shrink nature by a film carrying out orientation, and elongation increasing and on the other hand heat-treating polylactic acid further uniaxial stretching or by carrying out biaxial stretching is obtained. There is the description which does not sorb the component of a scent like cellophane as a description of the film which furthermore consists of a polylactic acid system polymer. For this reason, using a polylactic acid system film instead of the heat sealant material of the polyolefine system currently used from the former is expected. This is because it is dominance at the point which can be used in comfort to there being a

possibility that the sealant material of a polyolefine system may sorb the component of these scents depending on the object to pack, and contents may change.

[0006] When using as heat sealant material, it is raised that it is easy to laminate as a desirable property with that (1) seal temperature field is moderate, plastic film and paper besides (2), and a metallic foil, that shrinkage characteristics are low at the time of (3) seals, etc.

[0007] It is because it can pass that it can be used in a temperature region when the above (1) refers to the sealant material which consists of polyethylene used widely today as it is to the fabricating equipment used from the former, and the bag sealer specifically equipped with heat-sealing equipment, it does not newly need to introduce a facility and becomes economical. Although the seal temperature region of polyethylene is based also on thickness, even if it is high and low 130 degrees C or more, it is 80 degrees C or more, and it serves as a standard as heat sealant material. The above (2) shows that it is required to be able to laminate with the well-known equipment usually used by the dry laminate method and the wet laminating method. Lamination equipment \*\*\*\* one film, giving predetermined tension, after it applies heat if needed, it applies adhesives, and it is equipment which \*\*\*\* the film of another side, giving predetermined tension similarly, and piles up and sticks these two kinds of films by pressure through adhesives. therefore, tension -- or it is important that \*\*\*\* and an intermediate film does not fracture by delicate change of these tension and that a film dimension etc. does not change with the heat applied further.

[0008]

[Problem(s) to be Solved by the Invention] In the case of a polylactic acid system film, it is suitable to use the polylactic acid system film which carried out extension orientation from lamination fitness with a non-oriented film, as mentioned above, since it is weak. However, in order to obtain sufficient seal reinforcement which is used as a bag, for example, it is necessary to heat to temperature field extent which a film usually fuses but, the wrinkling by contraction, flapping, curl, etc. are looked at by the product which it contracted, and the oriented film did not satisfy the requirements for the above (3), but was done when the seal was carried out on such conditions, and if severe, it may not become a product at all.

[0009] Then, this invention aims at offering the oriented film of the polylactic acid system which has sufficient heat-resistant shrinkage characteristics while it has a heat-sealing property.

[0010]

[Means for Solving the Problem] The content rate  $D_a$  of D-lactic acid of the crystalline polylactic acid system polymer which this invention is a laminated film which uses a polylactic acid system polymer as a principal component, and which consists of two-layer at least, and constitutes one layer in the above-mentioned laminated film (%), The relation of the content rate  $D_b$  of D-lactic acid of the polylactic acid system polymer which constitutes other one layer of the above-mentioned laminated film (%)  $D_a \leq 7$  and -- It is  $D_b - D_a > 3$  and the above-mentioned technical problem was solved by making one layer besides the above constitute from one [ at least ] outermost layer of the above-mentioned laminated film.

[0011] Since the layer which consists of a crystalline polylactic acid system polymer which has predetermined D-lactic-acid content rate is made into one layer, it is hard to produce contraction deformation and heat-resistant shrinkage characteristics can be demonstrated. Moreover, since the layer which consists of a polylactic acid system polymer which has predetermined D-lactic-acid content rate is made into other one layer, it has sufficient heat-sealing property and the layered product obtained can be used as heat sealant material.

[0012]

[Embodiment of the Invention] Hereafter, the operation gestalt of this invention is explained.

[0013] The polylactic acid system laminating biaxial oriented film concerning this invention is a laminated film which uses a polylactic acid system polymer as a principal component and which consists of two-layer at least.

[0014] The above-mentioned polylactic acid system polymer is a polymer which comes to carry out condensation polymerization of the monomer which uses a lactic acid as a principal component. There are the L-lactic acid and D-lactic acid of two kinds of optical isomers among the above-mentioned lactic acids, and crystallinity differs at a rate of these two sorts of structural units. For example, the rate of L-lactic acid and D-lactic acid serves as transparency \*\*\*\*\* which there is no crystallinity and is softened near 60 degrees C of glass transition points by the random copolymer of

80:20-20:80 about. On the other hand, as for the random copolymer of 100:0-80:20, or 20:80-0:100, the rate of L-lactic acid and D-lactic acid has crystallinity about. Although that degree of crystallinity becomes it settled "The above-mentioned L-lactic acid and D-lactic acid be comparatively alike", the glass transition point of this copolymer is an about 60-degree C polymer like the above. After carrying out melting extrusion of this polymer, it becomes the ingredient of the amorphism nature which excelled [ quench / immediately ] in transparency, and serves as a crystalline ingredient by cooling slowly. For example, the homopolymer with which only L-lactic acid consists only of a D-lactic acid is a semicrystallinity polymer which has the melting point of 180 degrees C or more.

[0015] The polylactic acid system polymer concerning this invention is a polymer of D-lactic-acid unit and an L-lactic acid unit, may also include other hydroxycarboxylic acid units as a little copolymerization component, and may also contain little chain elongation agent residue.

[0016] As a polymerization method, well-known approaches, such as a condensation polymerization method and a ring-opening-polymerization method, are employable. for example, a condensation polymerization method -- if -- direct dehydration condensation polymerization of L-lactic acid, D-lactic acids, or such mixture can be carried out, and polylactic acid with the presentation of arbitration can be obtained.

[0017] Moreover, by the ring-opening-polymerization method (the lactide method), polylactic acid can be obtained using the selected catalyst, responding to \*\* the lactide which is the annular dimer of a lactic acid, and using a polymerization regulator etc.

[0018] As other above-mentioned hydroxycarboxylic acid units by which copolymerization is carried out to polylactic acid The optical isomer of a lactic acid (to L-lactic acid, it is L-lactic acid to D-lactic acid and D-lactic acid), A glycolic acid, 3-hydroxybutyric acid, 4-hydroxybutyrate, 2-hydroxy-n-butanoic acid, 2-hydroxy - Lactone, such as 2 organic-functions aliphatic series hydroxycarboxylic acid and caprolactones, such as 3 and 3-dimethyl butanoic acid, 2-hydroxy-3-methyl butanoic acid, 2-methyl lactic acid, and a 2-hydroxy caproic acid, a butyrolactone, and a valerolactone, is mentioned.

[0019] Moreover, hydroxycarboxylic acid other than non-aliphatic series diol like non-aliphatic series dicarboxylic acid and/or the ethyleneoxide addition product of bisphenol A like a terephthalic acid, and a lactic acid and/or a lactic acid may be used as a little copolymerization component if needed.

[0020] as the range where the weight average molecular weight of the polylactic acid system polymer used in this invention is desirable -- 60,000-700,000 -- it is -- more -- desirable -- 80,000-400,000 -- it is 100,000-300,000 especially preferably. If molecular weight is too small, practical use physical properties, such as machine physical properties and thermal resistance, will hardly be discovered, but when too large, melt viscosity is too high and inferior to fabrication nature.

[0021] One layer in the laminated film concerning this invention ("the 1st layer" is called hereafter.) consists of polylactic acid system polymers, and it is desirable that it is crystallinity. Moreover, other one layer ("the 2nd layer" is called hereafter.) of the above-mentioned laminated film consists of polylactic acid system polymers.

[0022] the content rate ("Da" is called below.) (%) of D-lactic acid of the crystalline polylactic acid system polymer which constitutes the 1st above-mentioned layer, and the content rate ("Db" is called below.) (%) of D-lactic acid of the polylactic acid system polymer which constitutes the 2nd above-mentioned layer --  $Da \leq 7$  and -- It is good to have the relation of  $Db - Da > 3$ .

[0023] That is, since the 1st layer turns into supporters, 7% or less of the rate (Da) of D-lactic acid in the crystalline polylactic acid system polymer which constitutes this 1st layer is desirable, and is more desirable. [ 5% or less of ] If the degree of crystallinity as supporters is low when it exceeds 7%, and thermal resistance is not obtained and is heated, it will be easy to carry out contraction deformation.

[0024] Moreover, since the 2nd layer turns into a heat-sealing layer, as for (Db), it is comparatively more desirable than Da that it is [ of D-lactic acid in the polylactic acid system polymer which constitutes this 2nd layer ] higher than 3%. It is because degree of crystallinity and the melting point will need to approach with the polylactic acid system polymer which constitutes the 1st above-mentioned layer and will need to carry out a seal at an elevated temperature, if this difference becomes 3% or less. That is, it is because supporters are also heated with a hot seal, a heat shrink happens, so the problem of lenticulating for a product and generating a wrinkling etc. is produced. Therefore, in order to lower degree of crystallinity and the melting point as compared with supporters, it is desirable to set it as the above-mentioned range.

[0025] In addition, the crystalline polylactic acid system polymer which constitutes the 1st

above-mentioned layer, and the polylactic acid system polymer which constitutes the 2nd above-mentioned layer may be mixtures of two or more kinds of different polylactic acid system polymers. In this case, D-lactic-acid rates Da and Db serve as the average computed from the blending ratio of coal of D-lactic acid which constitutes two or more kinds of polylactic acid system polymers, respectively.

[0026] Since the 2nd above-mentioned layer has a heat-sealing property, it constitutes one [ at least ] outermost layer of the above-mentioned laminated film.

[0027] The configuration of the laminated film concerning this invention has the 1st heat-resistant high layer in an interlayer, and 3 lamination of the 2nd layer / the 1st layer / the 2nd layer which consists of the 2nd layer which has a heat-sealing property to both sides is excellent in versatility. moreover, 5 lamination of the 2nd layer / the 1st layer / the 2nd layer / the 1st layer / the 2nd layer and the 2nd layer / the 1st layer / the 2nd layer / the 1st layer / ... /-- the 2nd-layer multilayer configuration may be used. moreover -- the 2nd layer [ from which only one side will serve as a heat-sealing layer if it takes into consideration to curl and thermal resistance of a film ] / 1st-layer two-layer configuration, or 4 lamination pan of the 2nd layer / the 1st layer / the 2nd layer / the 1st layer -- the 2nd layer / the 1st layer / ... /-- the 1st-layer multilayer configuration may be used. 2 micrometers or more of 5 micrometers or more of thickness of the 2nd layer which constitutes the outermost layer of these last multilayer films are 10 micrometers or more more preferably, and 10-100 micrometers of thickness of the last laminated film are 15-80 micrometers preferably.

[0028] Furthermore, thickness may carry out the laminating of the 10 micrometers or less of adhesives layers 5 micrometers or less, the resin layers for adhesion, recycle resin layers, or in-between layers (the 1st layer and the 2nd layer) preferably between each class between the 1st layer / the 2nd layer in the range which does not check the effectiveness of this invention.

[0029] Moreover, the crystalline polylactic acid system polymer which constitutes the 1st layer may be a mixture containing the polylactic acid system polymer which constitutes the 2nd layer, and may be recycle of all film layers.

[0030] In the polymer used by this invention, a thermostabilizer, light stabilizer, a light absorption agent, lubricant, a plasticizer, an inorganic filler, a coloring agent, a pigment, etc. can also be added in order to adjust many physical properties.

[0031] As the laminating approach, the approach used for usual is employable. For example, there are an approach of carrying out the so-called co-extrusion connected with one mouthpiece to a feed block system or a multi-manifold type and the approach of using the film of another kind and carrying out heating sticking by pressure of a roll or the press plate on the front face of the mixed film which it began to roll, from two or more extruders.

[0032] After quenching the sheet-like object or cylindrical object which extruded and carried out the polylactic acid system polymer from the T die, I die, the round-head die, etc. as the manufacture approach of the biaxial oriented film used as a principal component by the cooling cast roll, water, a compressed air, etc. and making it solidify in the near condition amorphously, the approach of extending to biaxial by the rolling method, the tenter method, tubular \*\*, etc. is mentioned.

[0033] Usually, in manufacture of a biaxial oriented film, it is the rolling method about vertical extension, and the biaxial extending method and the coincidence biaxial extending method extended by the tenter to coincidence in every direction for performing horizontal extension by the tenter method are common serially.

[0034] As extension conditions, it is a part for 100 - 10000%/preferably the extension temperature of 55-90 degrees C by two to 4 time, and 10 - 100000%/of extension rates two to 4 times, and 1.5 to 5 times as many horizontal draw magnification as this 65-80 degrees C and 1.5 times as many vertical draw magnification as this. However, since these fitness range changes with the presentation of a polymer, and heat histories of a non-extended sheet, it is decided suitably, taking the reinforcement of a film, and elongation into consideration.

[0035] When there is nothing in the range of the above-mentioned draw magnification and extension temperature, in the film by which falls remarkably and especially an extension postheat treatment is carried out, this inclination of the thickness precision of the obtained film is remarkable. Such a thickness deflection prints a film or becomes the factor which makes a product produce appearances, such as a wrinkling and flapping, severely in fabricating, such as bag manufacture, at a lamination pan with other film metallurgy group thin films and paper.

[0036] It heat-treats, where a film is grasped in the point which controls the heat shrink of a film. Usually, by the tenter method, since it is extended where a film is grasped with a clip, it heat-treats immediately. In the fabricating process of a film, it is easy to produce the problem of a film contracting during processing.

[0037] A thermostabilizer, light stabilizer, a light absorption agent, lubricant, a plasticizer, an inorganic filler, a coloring agent, a pigment, etc. can also be added in order to adjust many physical properties to above-mentioned each class.

[0038] As for contraction of the polylactic acid system laminating biaxial oriented film obtained, it is desirable that it is 5% or less after 80 degrees C of water baths and 1 minute. It is because a factor which makes a film produce appearances, such as a wrinkling and flapping, severely will arise if 5% is exceeded.

[0039] The polylactic acid system laminating biaxial oriented film manufactured by this invention turns into a polylactic acid system film which shrinkage characteristics were stopped as much as possible, and has heat-sealing nature. When using this biaxial oriented film as a packing material, and giving an example next, it is necessary to satisfy quality. For example, a backlining part, an insertion part, etc. are heat sealed for an individual package and a pyro package, and it is necessary to prevent from unpacking a package easily. Moreover, after heat sealing a film, manufacturing bags and putting the non-packing body into this bag, that opening may also be heat sealed and it may seal completely. In such a case, heat-sealing reinforcement needs to be 1Ns or more to film 15mm width of face. It is because a package will be cleared easily and the purpose of use will not be attained, if external force and an impact are added even if the seal reinforcement of a film is low, and it will heat seal and will make it a package object, if less than 1 N.

[0040] The polylactic acid system laminating biaxially oriented film concerning this invention Are suitable for the individual package for which a conventional biaxial-stretching polypropylene film and biaxial-stretching polyester film were used, a pyro package, and bag manufacture. And the polylactic acid system laminating biaxial oriented film of the biodegradability which can serve as a heat sealant layer when other plastic film is laminated can be offered. There is an application as a suitable film for the overlap of an individual package of confectionery, such as a fiber package, a vegetable package, chocolate, a caramel, and a candy, an audio tape, an audio disk, etc. Moreover, it can also be used, laminating with other plastic film, paper, aluminum foil, etc.

[0041]

[Example] Although an example is shown below, this invention does not receive a limit at all by these. First, the physical-properties measuring method in this example and the example of a comparison is shown in the following.

[0042] (1) Draw magnification and vertical draw magnification = the draw magnification of the flow rate and longitudinal direction of the original sheet before the flow rate / vertical extension of the film after vertical extension is the value which deducted the width of face of the part grasped to the clip of a tenter from the original sheet width of face before vertical extension, and is the value which assigned the die length which deducted the width of face of the part which was being grasped to the clip from the width of face obtained after horizontal extension.

Horizontal draw magnification = {(film width after extension) - (width of face which the clip was grasping)} / {(original sheet width of face before extension) - (width of face which the clip was grasping)}

[0043] (2) After starting die length of 100mm, and width of face to 100mm and being immersed in the 80-degree C warm water bus for 1 minute along with MD and TD of a contraction film, the dimension after the contraction was measured and the rate of a heat shrink was computed according to the degree type. Contraction is measured along the extension direction and MD and TD serve as the trial direction in an exam.

Contraction (%) = {(dimension before contraction) - (dimension after contraction)} x 100 / (dimension before contraction)

[0044] (3) The film cut down to heat-sealing reinforcement and result MD170 mm x TD130mm was prepared, the seal was carried out with superposition and a heat-sealing bar, and the two-way-type seal bag which the base met and inserted into MD was produced so that the fields which fold this in a two way type and serve as heat sealant material might contact. Seal conditions were cooled radiationally after pressing for about 5 seconds at 80, 100 and 120 degrees C. the width of face of a heating bar -- 10mm

and pressure 1.5 kgf/cm<sup>2</sup> it is .

[0045] The bag was observed after the seal and a wrinkling, flapping, contraction unevenness, etc. wrote it as x to what is seen remarkably. although these are seen a little on the other hand -- enough -- as a bag -- finish -- \*\*\*\* -- about what is, it was written as O. Heat-sealing strength is JIS. It asked for the reinforcement in 15mm width of face by the approach indicated by Z1711 "polyethylene film bag manufacture." Therefore, one end was made into Shilu, and the film was cut down in width of face of 15mm so that it might become perpendicular to the longitudinal direction of the seal (getting it blocked and meeting MD of a film). It examined by opening the cut-down film and grasping by the chuck of a tension tester. The bad bag of workmanship of seal reinforcement is also comparatively bad, and tends to vary. About what the film contracted remarkably, it could not measure but it was written that measurement was impossible.

[0046] (4) All evaluations of contraction of the comprehensive evaluation above, heat-sealing reinforcement, and a result made O thing O, and made x the thing with at least one x.

(Configuration resin of a layered product) As resin which constitutes a layered product, the mixture of the 1st component independence shown in Table 1 or the 1st component, and the 2nd component was used. D-lactic-acid rate in the case of a mixture was computed as the average from both weight fraction.

[0047]

[Table 1]

樹脂番号	1	2	3	4	5	6
平均D乳酸	0.5 %	5 %	8 %	9 %	10%	17%
第1成分 D乳酸割合	0.5 %	5 %	8 %	10 %	10 %	20 %
ガラス転移点	58 ℃	56 ℃	53 ℃	52 ℃	52 ℃	52 ℃
分子量	24 万	20 万	18 万	18 万	18 万	19 万
第2成分 D乳酸割合				5 %		5 %
ガラス転移点				56 ℃		56 ℃
分子量				20 万		20 万
第1成分割合	100%	100%	100 %	80 %	100 %	80 %
第2成分割合				20 %		20 %
結晶性						

[0048] It has the structural unit of D-lactic-acid =80:20. L-lactic acid : 80% of glass transition point (Tg)52 degree C polylactic acid, (Example 1) L-lactic acid : Have the structural unit of D-lactic-acid =95:5 and 20% of glass transition point (Tg)56 degree C polylactic acid is mixed. With the with a mean particle diameter of 1.4 micrometers dried to polylactic acid (resin 6 of Db=17% and table 1) of a total of 100 weight sections said direction twin screw extruder which granular silica (trade name: SAIRISHIA 100, product made from Fuji SHIRISHIA Chemistry ) 0.1 weight section mixing is carried out, and is 25mmphi It extruded as a table lining from the mouthpiece of a multi-manifold type at 220 degrees C.

[0049] moreover, the structural unit of L-lactic acid:D-lactic-acid =99.5:0.5 (Da=0.5%) -- having -- a glass transition point (Tg)58 degree C polylactic acid polymer (resin 1 of Table 1) -- 40mmphi single screw extruder -- the above -- it extruded as an interlayer from the mouthpiece.

[0050] The thickness ratio of a surface, an interlayer, and a lining adjusted the discharge quantity of melting resin so that it might be set to 1:10:1. This co-extrusion sheet was quenched with about 43-degree C casting roll, and the non-extended sheet was obtained. then, a longitudinal direction -- 76

degrees C -- 2.6 times as many roll extension as this -- subsequently to [ 3.2 times ], it extended by the tenter crosswise and extended at the temperature of 72 degrees C. Temperature of the heat treatment zone in a tenter was made into 130 degrees C, and produced the heat-treated film. Film thickness adjusted the discharge quantity and line speed of melting resin from an extruder so that it might be about set to 30 micrometers on an average. The evaluation result of a film is shown in Table 2.

[0051] (Examples 2 and 3 and examples 1-5 of a comparison) As shown in Table 2 or 3, the biaxial extension postheat treatment was extruded and carried out, and the film was produced so that the polylactic acid system polymer (it is equivalent to each resin given in Table 1.) with which L-lactic acid differs from D-lactic acid might be made respectively like an example 1, it might be made a surface, an interlayer, and a lining and it might become a predetermined thickness ratio. The evaluation result of each film is shown in Table 2 and 3.

[0052] In addition, the example 2 of a comparison fractured the film during heat treatment. In the examples 1 and 4 of a comparison, the appearance of the produced bag which carried out the seal was bad, and was not able to obtain the good thing of workmanship. Moreover, the examples 3 and 5 of a comparison were not able to obtain sufficient seal reinforcement which can take a gestalt as a bag. The evaluation result of each film is shown in Table 2 or 3.

[0053] It has the structural unit of D-lactic-acid =80:20. L-lactic acid : 80% of glass transition point (Tg)52 degree C polylactic acid, (Example 4) L-lactic acid : Have the structural unit of D-lactic-acid =95:5 and 20% of glass transition point (Tg)56 degree C polylactic acid is mixed. With the with a mean particle diameter of 1.4 micrometers dried to polylactic acid (resin 6 of Db=17% and table 1) of a total of 100 weight sections said direction twin screw extruder which granular silica (trade name: SAIRISHIA 100, product made from Fuji SHIRISHIA Chemistry ) 0.1 weight section mixing is carried out, and is 25mmphi It extruded at 210 degrees C as a surface from the mouthpiece of a two-layer multi-manifold type.

[0054] moreover, granular silica (trade name: SAIRISHIA 100, product made from Fuji SHIRISHIA Chemistry ) 0.1 weight section mixing with a mean particle diameter of 1.4 micrometers which had the structural unit of L-lactic acid:D-lactic-acid =99.5:0.5 (Da=0.5%), and was dried to the glass transition point (Tg)58 degree C polylactic acid polymer (resin 1 of Table 1) -- carrying out -- 40mmphi single screw extruder -- 210 degrees C -- the above -- it extruded as an interlayer from the mouthpiece. Since the layered product obtained is two-layer structure, an interlayer forms a lining as it is.

[0055] the thickness ratio of this layered product -- surface: -- the discharge quantity of melting resin was adjusted so that an interlayer might be set to 1:2. This co-extrusion sheet was quenched with about 42-degree C casting roll, and the non-extended sheet was obtained. then, a longitudinal direction -- 75 degrees C -- 2.6 times as many roll extension as this -- subsequently to [ 3.6 times ], it extended by the tenter crosswise and extended at the temperature of 74 degrees C. Temperature of the heat treatment zone in a tenter was made into 135 degrees C, and produced the heat-treated film. Film thickness adjusted the discharge quantity and line speed of melting resin from an extruder so that it might be about set to 15 micrometers on an average. The evaluation result of a film is shown in Table 2.

[0056]

[Table 2]

実施例		1	2	3	4
構成		3 層	3 層	3 層	2 層
表層の D-乳酸割合 (D b) (%)		17	9	10	17
中間層の D-乳酸割合 (D a) (%)		0.5	0.5	5	0.5
裏層 D-乳酸割合 (D b) (%)		17	9	10	
D b - D a (%)		16.5	8.5	5	16.5
フィルム全厚み (μm)		30	40	40	15
厚み比 (第 1 層 / 第 2 層 / 第 3 層)		1/10/1	1/2/1	1/2/1	1/2
縦延伸温度 (℃)		76	76	78	76
横延伸温度 (℃)		72	72	74	72
縦延伸倍率		2.6	2.6	2.9	2.6
横延伸倍率		3.2	3.2	4.5	3.2
熱処理温度 (℃)		130	130	127	135
熱処理時間 (sec)		15	20	18	10
80℃/1分間の収縮率 (%)	MD	2	3	4	3
	TD	2	2	3	2
ヒートシール強度 および仕上がり (N/15mm)	80℃	3.0 ○	0.0 ○	1.1 ○	5.9 ○
	100℃	3.0 ○	11.2 ○	8.8 ○	4.7 ○
	120℃	4.1 ○	7.2 ○	7.4 ○	5.3 ○
備考					
総合評価		○	○	○	○

[0057]

[Table 3]

比較例		1	2	3	4	5
構成		3層	3層	3層	3層	3層
表層のD-乳酸割合 (D <sub>b</sub> ) (%)		17	17	8	8	0.5
中間層のD-乳酸割合 (D <sub>a</sub> ) (%)		8	8	5	5	17
裏層D-乳酸割合 (D <sub>b</sub> ) (%)		17	17	8	8	0.5
D <sub>b</sub> - D <sub>a</sub> (%)		9	9	3	3	16.5
フィルム全厚み (μm)		40	40	40	40	30
厚み比 (第1層/第2層/第3層)		1/2/1	1/2/1	1/2/1	1/2/1	1/1/1
縦延伸温度 (℃)		76	76	78	78	76
横延伸温度 (℃)		74	74	74	74	72
縦延伸倍率		3.1	3.1	2.9	2.9	2.6
横延伸倍率		4.5	4.5	4.5	4.5	3.2
熱処理温度 (℃)		110	130	125	90	130
熱処理時間 (sec)		16	16	18	18	15
80℃/1分間の収縮率 (%)	MD	8	評価 できず	4	32	2
	TD	6		3	35	1
ヒートシール強度 および仕上がり (N/15mm)	80℃	4.4 ○	評価 できず	0.5 ○	測定不能 ×	0 ○
	100℃	測定不能 ×	評価 できず	0.3 ○	測定不能 ×	0.1 ○
	120℃	測定不能 ×	評価 できず	0.2 ○	測定不能 ×	0.3 ○
備考		仕上り 不良	フィルム 破断	シール できず	仕上り 不良	シール できず
総合評価		×	×	×	×	×

[0058]

[Effect of the Invention] The layered product by this invention has the supporters who have heat-resistant shrinkage characteristics, and the heat-sealing layer which has a heat-sealing property.

[0059] Moreover, the layered product by this invention can offer the polylactic acid system laminating biaxial oriented film of the biodegradability used as a heat sealant layer, when other plastic film is laminated suitable for the individual package for which a biaxial-stretching polypropylene film and biaxial-stretching polyester film were used conventionally, a pyro package, and bag manufacture.

[Translation done.]